Some remarks on simultaneous measurements of particulate contaminants including radioactivity and isotopic composition of precipitation

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ABSTRACT

Results of simultaneous measurements of trace substances in rain sampled during the passage of precipitation systems are discussed. The pattern of the concentrations for particulate matter is compared with that for the O^{18} and deuterium isotopes. The variations in the concentration have been investigated for various synoptic situations.

1. Introduction

A number of authors discussed during recent years data on the variability of the concentrations of radioactive and other trace substances during rain- or snowstorms, in order to contribute to the clarification of the mechanism that cleans the atmosphere from contaminants (BLEICHRODT *et al.*, 1959; GEORGII, 1962; HUFF & STOUT, 1964; BLEEKER & LABLANS, 1965). Data on the variability of the isotopic composition of water during precipitation events were also presented (DANSGAARD, 1953, 1961, EPSTEIN, 1956, EHHALT et al. 1963).

It was noted that the nature of the variations in both groups of investigations was more or less similar. This was a motive for the present authors to combine the measurements of radioactive and other trace substances of particulate origin with those of the isotopic composition of the same precipitation samples in order to investigate the simultaneous behaviour of the various components.

Results are now discussed on the basis of current ideas on the scavenging mechanism as reviewed recently by BLEEKER & LABLANS (1965) and on ideas about the factors which control the isotopic composition of precipitation as discussed by DANSGAARD (1964).

2. The scavenging of particulate matter from the atmosphere

2.1. *Collection of the data*

During the period from 12 July 1961 to 22 January 1965 approximately 30 cases of rain- or snowstorms (showers and frontal passages) were sampled in such a way that for every storm a sequence of data on the concentration of some constituents (gross-radioactivity, Na^+-, Cl^--, Ca^{++}-ions) became available. Also the conductivity was measured as an overall information on the ion content. The aim of the investigation was to check the at its beginning still scanty and sometimes contradictory information on the mechanism(s) that scavenge(s) the atmosphere from radioactive and other particulate matter.

The experimental procedure and some of the results have been discussed elsewhere (BLEEKER & LABLANS, 1965).

2.2. *Scavenging for a simple model of a precipitation system*

BLEEKER & LABLANS (1965) indicated that it appears possible to give a meteorological interpretation of their data on the basis of the work of GREENFIELD (1957) who showed that particulate matter of aerosol-dimensions is essentially scavenged from the atmosphere by the coalescence of aerosol-particles and cloud droplets owing to collisions caused by the Brownian motion of the particles and the turbulent motion of the air. The results obtained may be summarized best by describing what seems to be typical for a simple model of a precipitation system and by discussing some anomalies.

We define a simple precipitation system as a precipitating cloud system, that develops

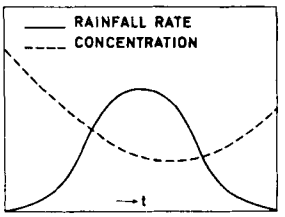

FIG. 1. Variation of the concentration of a particulate contaminant during the passage of a simple precipitation system.

in an airmass which is horizontally homogeneous from a chemical point of view, that has a one-topped precipitation rate recording and that is preceeded and is followed by broken or clear sky. BLEEKER and LABLANS (1965) concluded that the concentrations of any contaminant usually show a maximum at the beginning and at the end of the storm, the first maximum being the highest and that the minimum in between coincides approximately with the maximum in the rainfall rate (Fig. 1).

In order to explain the observed behaviour of the concentrations of the contaminants it is not sufficient to consider the primary scavenging mechanism, indicated above. It is in addition necessary to recognize the meteorological parameters related to this primary mechanism. As such the following may be mentioned: the lifetime of the cloud droplets, the liquid water content of the cloud, the precipitation rate, the height of the cloud-base, the evaporation of cloud droplets and the evaporation of precipitation elements.

These factors are of course strongly intercorrelated. As an example, a high precipitation rate will stem from a part of the cloud system with a relatively large amount of liquid water, where the cloud droplets may have only a relatively short lifetime. It will be shown in the next subparagraph that this combination of circumstances leads to low concentration of the contaminants in the precipitation. Moreover, evaporation which always leads to augmentation of the concentration of the contaminants will under those circumstances be of little importance, since the cloud-base will be low and the air under the cloud-base will be moist. This explains why the minimum in the concentration of the contaminants falls approximately at the same time as the maximum in the precipitation rate (Fig. 1).

The situation is quite different during the period of the rainstorm in advance of or after the period of high rainfall rate. The rain of low intensity falls from parts of the cloud system with a lower water content (ZAITSEV, 1950), where the cloud droplets have relatively long lifetimes. The cloud-base will be higher so that the, usually smaller, precipitation elements have to pass a thick layer of non-saturated air which leads to considerable evaporation (MASON, 1952). Moreover at the edges of the cloud system the concentration of the contaminants in the cloud droplets will be augmented as a result of evaporation of the droplets caused by the entrainment-process. In addition, it is to be expected that the contaminant concentration in the cloud droplets will be high as a result of the fact that the air introduced in the cloud has the full concentration of the concerned contaminant, or in other words this air has not yet been depleted from contaminants as a result of Brownian and turbulent motion.

Thus the high values of the concentration at the beginning and the end of the rainstorm are explained. The fact that the maximum at the end of the rain is generally lower than the maximum at the beginning may be considered as resulting from a gradual increase of the humidity of the air under the cloud-base during the passage of the system.

2.3. *Deviations of actual cloud systems from the simple model*

It is sometimes observed that a temporarily increase of the rainfall rate is accompanied by an *increase* in the gross-radioactivity, while other constituents show the "normal" behaviour as described above. This anomaly is explained by assuming that in the uppermost parts of the clouds air can be entrained from the upper-tropospheric or stratospheric layers, which had a high content of radioactive particles (KRUGER *et al.*, 1963; BLEEKER & LABLANS, 1965). Kruger *et al.* consider this phenomenon as typical for Cumulonimbus-clouds. The phenomenon showed up in our measurements in rainstorms originating from convective as well as from layered frontal clouds; it is apparently related to the accidental presence of high radioactivity in the upper-tropospheric layers.

It was already indicated that the minimum in the observed concentrations coincides approximately with the maximum in the rainfall

rate. From a more detailed inspection of the data it appears that there exists a time shift; the concentration-minimum is always observed somewhat later than the rainfall rate maximum. This time shift could be explained as resulting from the not symmetrical action of the evaporation under the cloud-base in the model, the air under the cloud-base becoming increasingly moister with the passing of the system.

The shift could, however, also be considered as one of the anomalies for which the model does not allow. The explanation is then based (BLEEKER & LABLANS, 1965) on the fact that in actual cloud systems the windspeed usually increases with height. The upper part of the updraughts will therefore be shifted towards the leeside of the cloud. Rain from the most leeward updraughts will then, on its way down, pick up cloud particles from the more peripheral parts of the cloud which have a high content of contaminants due to their long residence time and possibly to entrainment effects. Rain from later updraughts will consist entirely of cloud droplets of a short residence time. Thus, the lowest values of the concentration of the contaminants may only be expected some time after the onset of the heavy precipitation.

Sometimes the concentration of a contaminant is higher at the end than at the beginning of a rainstorm, contradictory to what is expected for the model. This may occur when the contamination of the air is high in the rear of a cloud system which accompanies a front. A case study of this phenomenon with respect to gross-radioactivity has been presented by BLEICHRODT, BLEEKER & SCHMIDT (1960).

2.4. *The theory of Greenfield and the observed concentrations of particulate contaminants in precipitation.*

GREENFIELD (1957) gives an expression for the number of particles per cm³, $N(t)$, of radius r_p, that remain in air which originally has a content of N_0 particles per cm³, after collection of these particles by a cloud consisting of n_c cloud droplets, with radius r_c, per cm³ during a period of t, namely:

$$N(t) = N_0 e^{-Kn_c t} \qquad (1)$$

In this equation K is a function of r_p, r_c and the velocity gradient ω normal to the streamlines. K represents the influence of Brownian motion and turbulence on the coagulation of particles and cloud drops.

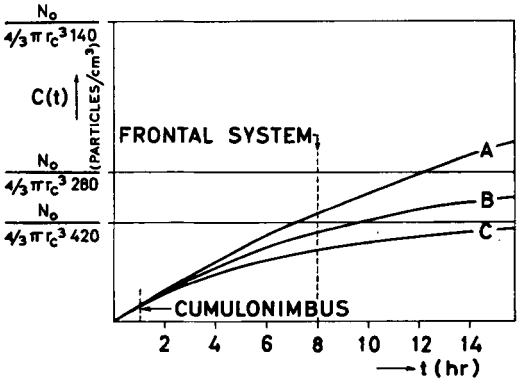

FIG. 2. Concentration of particulate matter in the cloud water versus time for various numbers n_c of cloud droplets per cm³. Curve A: $n_c = 140$, Curve B: $n_c = 280$, Curve C: $n_c = 420$.

The values of n_c and r_c determine the liquid water content of the cloud:

$$\lambda = \tfrac{4}{3}\pi r_c^3 n_c. \qquad (2)$$

For the interpretation of our measurements of the concentrations of radioactive and other particulate matter in the precipitation samples it is useful to transform equations (1) and (2) into an expression for the concentration $C(t)$ of particulate matter in cloud water as a function of time

$$C(t) = N_0(1 - e^{-Kn_c t})/\tfrac{4}{3}\pi r_c^3 n_c. \qquad (3)$$

In Fig. 2 this expression is plotted for aerosol-particles of radius 0.1 μ that are scavenged by cloud droplets of radius 15 μ. The curves A, B and C correspond respectively to $n_c = 140$ ($\lambda = 2$ gr/m³), $n_c = 280$ ($\lambda = 4$ gr/m³) and $n_c = 420$ ($\lambda = 6$ gr/m³). $\omega = 20$(cm/sec)/cm; it is thought that these values are not unreasonable assumptions for precipitating cumulonimbus as well as for precipitating frontal systems. (The value of $\lambda = 0.4$ gr/m³ used by GREENFIELD (1957) presumably originates from data obtained from non-precipitating stable stratus.)

In a frontal system the droplets may reside long enough to collect, in the denser parts, an important portion of the aerosol particles. Fig. 2 shows that, owing to the presence of many cloud droplets, the scavenging of the aerosol in the dense parts of the cloud is relatively rapid,

but also that owing to their competition the concentration of the contaminant in the cloud water always remains lower than in the less dense regions since the particles are divided over a relatively large mass of water. This is very clear from the limiting value of $C(t)$:

$$C(\infty) = N_0/\tfrac{4}{3}\pi r_c^3 n_c \qquad (4)$$

which indicates that for very long collection times the concentration of the contaminant is inversely proportional with the number of cloud droplets.

For the short lived cumulonimbus we find, owing to t being of the order of less than one hour, only low figures for the scavenged fraction.

For these low figures the concentration of the contaminant in the cloud water is essentially insensitive to variations of n_c which may be seen when we replace $1 - \exp(-Kn_c t)$ by $Kn_c t$ in (3), leading to

$$C(t) = (N_0 K/\tfrac{4}{3}\pi r_c^3)t. \qquad (5)$$

This indicates that the collection efficiency of the cloud droplets before their coagulation to precipitation elements is not seriously affected by the competition of their neighbours.

Attention should be drawn to the possibility. that there will exist a relation between the value of n_c and the residence time of cloud droplets in precipitating clouds. In stratiform as well as in cumuliform precipitating cloud systems the residence time is presumably shorter if the number of cloud droplets is larger and vice versa. If we assume that the rainfall rate is positively correlated to the liquid water content in the region of formation, then we may conclude that the scavenging theory of Greenfield predicts for stratiform precipitating clouds (t and n_c are the governing factors) and for cumuliform precipitating clouds (t is the governing factor) the inverse relation between the concentration of the contaminants and rainfall rate, as reported by BLEICHRODT *et al.* (1959), GEORGII (1962), BLEEKER & LABLANS (1965) and others.

3. The isotopic composition of precipitation

3.1. *Introduction*

The processes governing the concentration of the isotopic water components in cloud- and precipitation elements are in many respects quite different from those treated above. Simultaneous consideration of chemical and isotopic composition of the precipitation during a rain- or a snowstorm may nevertheless shed some new light on the processes involved.

Several authors contributed to our present knowledge about the isotopic composition of natural waters (Craig, Ehhalt, Epstein, Facy *et al.*, Friedman *et al.*, Gonfiantini *et al.* and DANSGAARD, 1964). The latter paper gives the references and a more detailed review on the matter than space allows here; therefore, only the main points of interest to the present investigation are briefly given below.

The heavy istope concentration, δ, is always expressed in relative units, namely as its per mille difference from that of standard water (SMOW).

Of primary importance for the concentration of the isotopes is the exchange process between the liquid and the vapour phase, which is governed by the so-called fractionation factor α, defined by

$$\alpha = \frac{(a_{\text{light}}/a_{\text{heavy}})\ \text{vapour}}{(a_{\text{light}}/a_{\text{heavy}})\ \text{liquid}},$$

where a is the absolute content of the components in the mixtures.

From Raoult's law which applies to equilibrium conditions it appears that

$$\alpha = \frac{p_{\text{light}}}{p_{\text{heavy}}},$$

where p is the saturation vapour pressure of the various isotopic components. At normal temperature the ratio's of the p values for the most important components are:

$$p(H_2O^{16})/p(H_2O^{18}) = 1.01$$

and

$$p(H_2O)/p(HDO) = 1.08$$

This means that under equilibrium conditions vapour is depleted approximately 10.0 ‰ in O^{18} and 80.0 ‰ in deuterium relative to the liquid.

Consequently in an evaporating liquid the evaporation process causes an increase of the δ-value of the remaining liquid phase as a result of the loss of lighter molecules.

On the other hand during a condensation process, i.e. in cooling moist air, the δ's in the

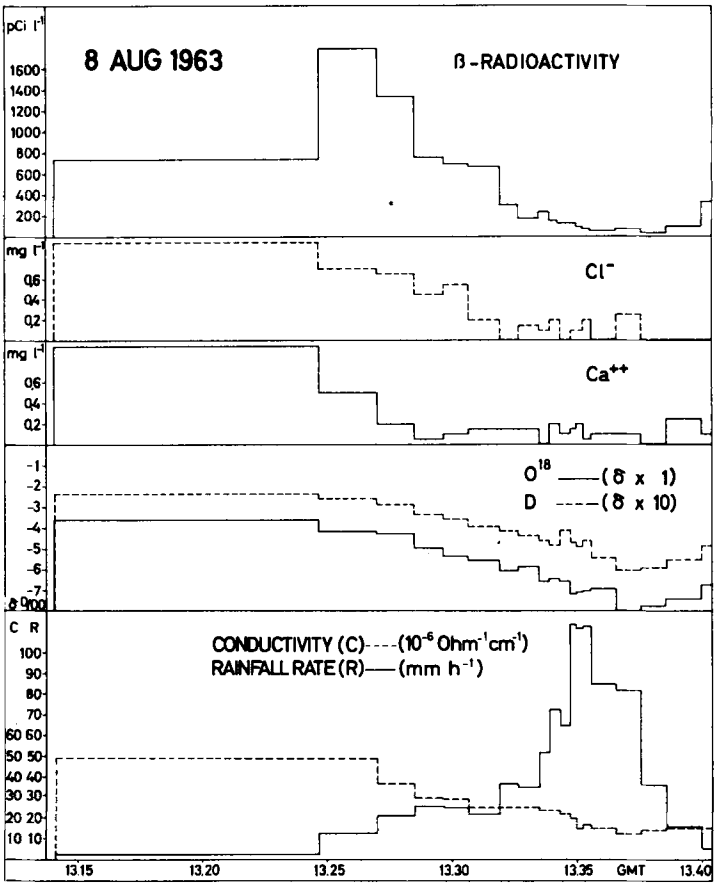

Fig. 3*a*. Passage of a shower on 8 August 1963. Rainfall rate, conductivity, deuterium, oxygen-18, Ca^{++}, Cl^-, and specific radioactivity versus time.

remaining vapour will decrease as a result of the preferential condensation of the heavy component. Also the δ in the condensate will decrease; this may be understood by realizing that the δ-value of the first condensate is relatively high.

In both processes, i.e. in the evaporation and the condensation, the changes in the δ are highest if the newly formed phase is removed from the system.

If for an equilibrium (slow) evaporation-condensation process, δ_D is plotted against δ_{18}, one finds a linear relation between δ_D (ordinate) and δ_{18} (abscissae) with a slope of 8. This slope will be less than 8 in case of non-equilibrium (fast) *evaporation* (in nature always combined with exchange with environmental vapour e.g. in falling raindrops); this is partly due to the fact that HDO^{16} evaporates slower than H_2O^{16} but faster than H_2O^{18}. Non-equilibrium *condensation* has never been proved to occur in nature, probably because the vapour pressure gradients in a condensation process are much smaller than those usually occurring in evaporation processes.

It appears from theoretical considerations (FRIEDMAN, 1962) that complete equilibration between cloud droplets and vapour is a matter of only a few seconds. However the equilibration between raindrops and vapour takes several minutes, depending on the drop size; this is the reason why the δ's of rain partly reflect the conditions in the cloud and not only those below the cloud-base.

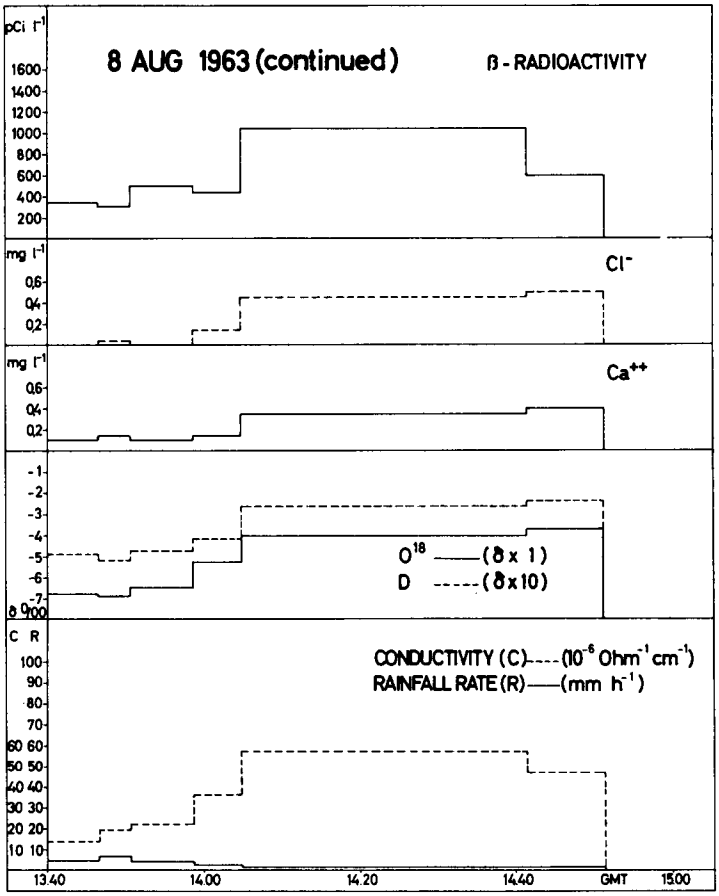

FIG. 3*b*. Passage of a shower on 8 August 1963, continued.

3.2. *The isotopic composition of the rain for a simple model of a precipitation system*

Let us first consider the centre of our model precipitation system as outlined in sub-paragraph 2.2. Ascending air has been cooled moist adiabatically; the condensation process started at the cloud-base. As explained in sub-paragraph 3.1 it may be expected that the δ's of both phases decrease in vertical direction.

The δ_{18}- and the δ_D-values of a small raindrop which started falling from the higher parts of the cloud are initially low. On its way down in the cloud, the rain-drop will collect cloud droplets with higher δ-values. Furthermore, on its way from the cloud-base to the ground, there are two processes to be taken into consideration, namely the exchange process between the drop and the water vapour environment with higher values of δ and the evaporation of the drop which will take place if the drop is not too cold. Both processes lead to further increase of the δ-values. The pure evaporation process under non-equilibrium conditions would, when the δ's of various samples are plotted in a δ_{18}–δ_D-diagram, give a line with a slope < 8.

In case low values of δ's are found they must be due to one of the following factors: low initial δ of the vapour, high degree of adiabatic cooling or relatively little effect of collection of cloud droplets in the lower layers and of exchange and evaporation processes. Since the degree of adiabatic cooling is higher and the influence of exchange and evaporation is lower in the centre of the cloud relatively low δ's may be expected there.

In the marginal zones and especially in the

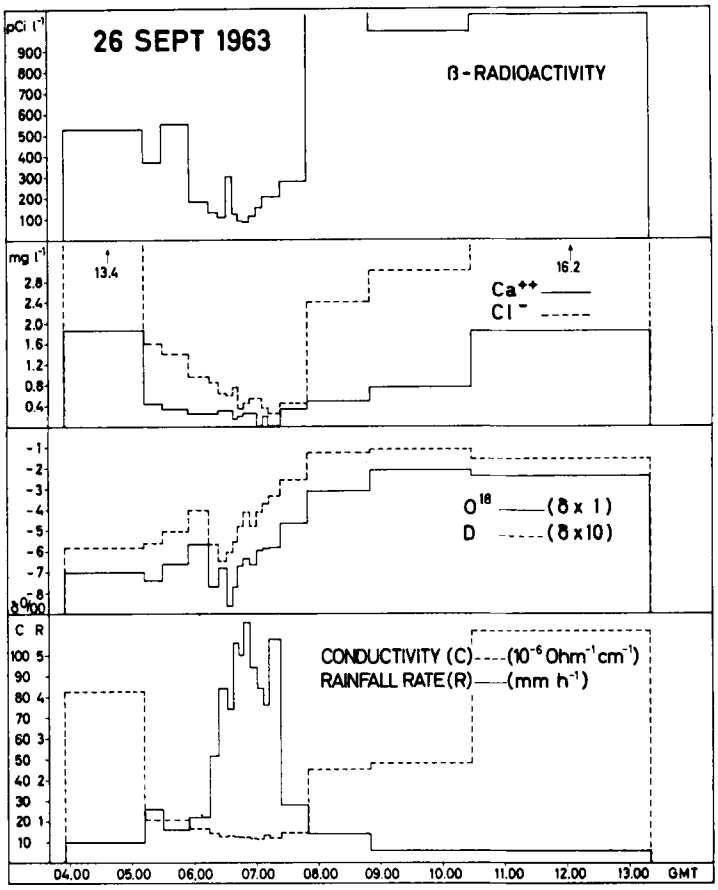

FIG. 4. Passage of a warmfront on 26 September 1963. Rainfall rate, conductivity, deuterium, oxygen-18, Cl^-, Ca^{++} and specific radioactivity versus time.

foreward part of the precipitation system we must expect higher values of the δ's of the rain, firstly because part of the rain will originate from air which has not risen so high as the air in the centre, and secondly because the evaporation under the cloud-base of the smaller raindrops is larger. We may therefore expect that the registrations of the δ-values as a function of time have essentially the same shape as those for the concentration of particulate matter.

In the next paragraph we will investigate how far this analogy was realised in some of the actual precipitation systems.

4. Discussion of some simultaneous series of measurements of particulate contaminants and of the isotopic composition of precipitation

4.1. *Shower*

Fig. 3 demonstrates the data as obtained from a rain-shower, passing De Bilt on 8 August 1963. The synoptic situation is indicated on Fig. 8.

The shower behaves as a model precipitation system. This is in accordance with the relative simplicity of the meteorological situation: an isolated shower that developed at the passage

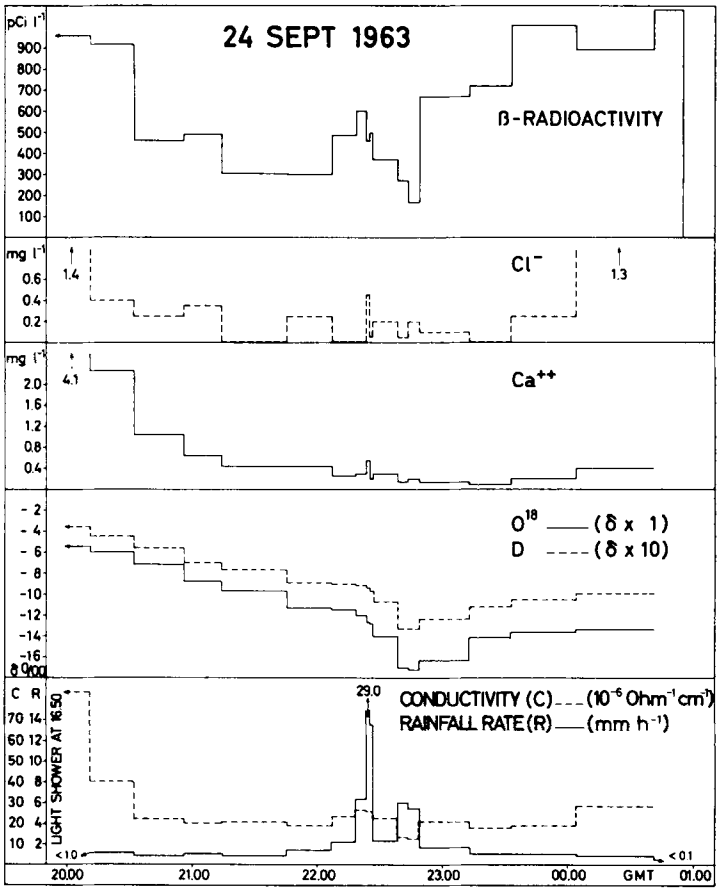

FIG. 5. Passage of a coldfront on 24 September 1963. Rainfall rate, conductivity, deuterium, oxygen-18, Ca^{++}, Cl^- and specific radioactivity versus time.

of a weak cold front (cf. DANSGAARD, 1961, p. 70).

We notice that the minima of the concentration of the contaminants and of the δ's all have the same time-shift (of the order of a few minutes) relative to the precipitation rate. The first explanation for the time-shift in the curves for particulate matter, namely the influence of the gradually decreasing evaporation, may also explain the time-shift of the δ's, since decreasing evaporation means that low δ's will still occur after the passage of the most intensive rainfall.

4.2. *Warmfront*

The rain of 26 September 1963 (fig. 4) is already of higher complexity. The meteorological difference between this system and the model system is that the present system refers to a warmfront (Fig. 8). The behaviour of the particulate constituents does not deviate much from that in a model system, although it is apparent that the values at the end of the system are slightly higher than those at the beginning.

As the warmfront passes De Bilt the rain will originate from lower and lower altitudes, i.e. from air which has been exposed to less and less cooling. This corresponds to a general rise in the δ's. Such a trend is recognized in figure 4, where it is superimposed upon the model response to the rate of rainfall. This pattern is very much like that found by DANSGAARD, (1953) in a similar situation.

4.3. *Coldfront*

Fig. 5 gives the data collected on 24 September 1963 when a coldfront passed De Bilt (for synoptic situation see Fig. 8).

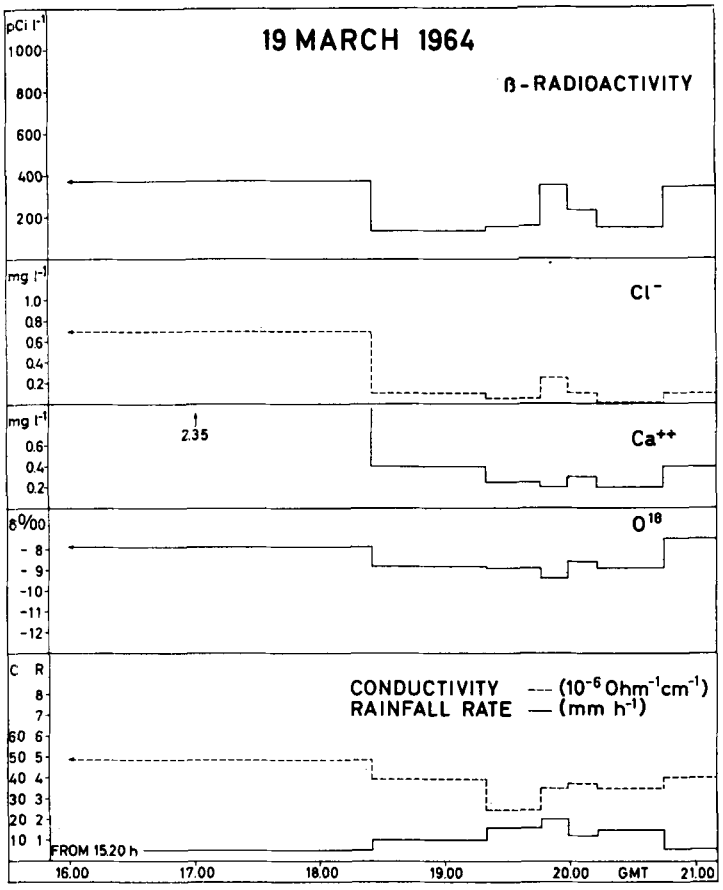

FIG. 6*a*. Passage of a complex frontal system on 19 March 1964. Rainfall rate, conductivity, oxygen-18, Ca^{++}, Cl^{-}, and specific radioactivity versus time.

For the concentration of particulate contaminants the picture of the simple model can be recognized. There exists however an interesting deviation, demonstrated by anomalous high values in the radioactivity at the peak of the rainfall rate about 22.25 G.M.T., when apparently air with high specific activity was entrained in the cloud system, presumably in its upper part. The second peak in the rainfall rate at 22.45 G.M.T. was not accompanied by high values of the radioactivity; on the contrary the values were low which is to be expected if no air of the higher layers is entrained.

With respect to the δ-values, it is interesting to see that in this coldfront-case a trend opposite to that in the warmfront is clearly present, as indicated by the general descent.

4.4. *Complex frontal system*

The data obtained on 19 March 1964 at De Bilt, given in figure 6, look rather complicated. The synoptic situation represented in fig. 8, shows a weak double warmfront with rain from 16.00–21.00 G.M.T., followed by a coldfront occlusion with rain from 21.00–02.00 G.M.T.

The particulate contaminants show an anomaly at 20.00 G.M.T., when radioactivity, chlorine and the overall ion content suddenly rise considerably with a slight increase of the rainfall rate. Presumably subtropical air of maritime origin with a high chlorine content, low calcium content (GAMBELL, 1962) and also a higher value of the radioactivity takes part

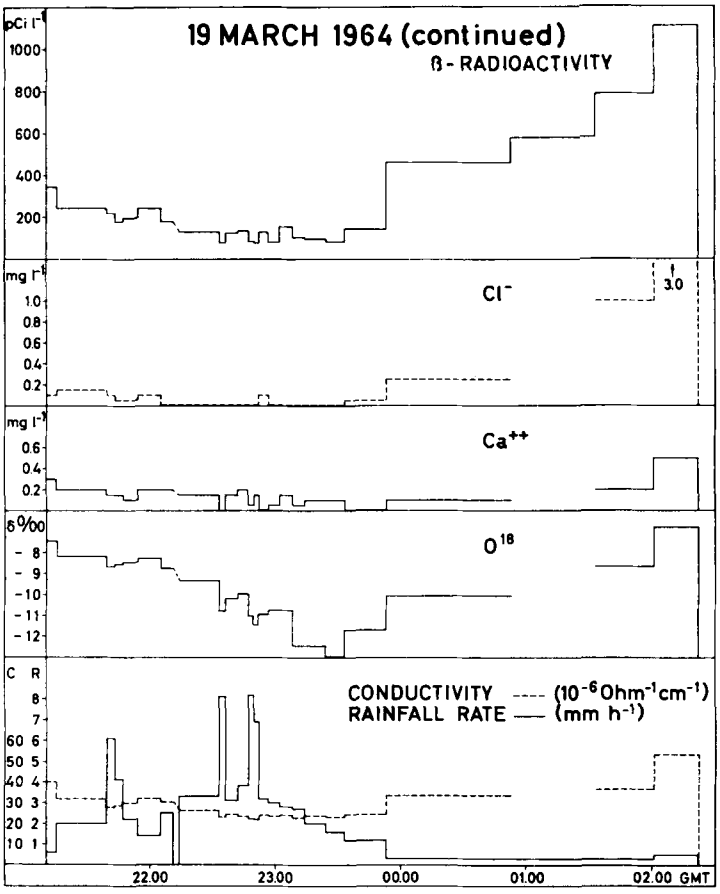

FIG. 6b. Passage of a complex frontal system on 19 March 1964, continued.

in the precipitation process of the double warmfront system.

The data of δ_{18} can be considered as a response to increasing rainfall rate in combination with a slight rise corresponding to the warmfront character of the rain (until 21.00 G.M.T.), followed by a strong fall, in excellent agreement with the coldfront character of the rain.

4.5. *Frontal system*

The rain of 24 March 1964 at De Bilt (Fig. 7 and Fig. 8) can also be divided into two parts, which are separated by a period of low rainfall rate at about 19.00 G.M.T. In the first section the response of radio-activity, calcium, conductivity and the δ-values to the temporarily low rainfall at about 15.00 G.M.T. strikes the eye. The particulate contaminants remain until 18.00 G.M.T. on levels that are in good accordance with the rainfall rate. The δ's even follow the rainfall rate in close correlation between 13.00 and 16.00 G.M.T. (intensity effect, subparagraph 3.2); after 16.00 G.M.T. they show a general rise corresponding to the warmfront character of the rain during that period. At 18.00 G.M.T. the concentrations of the contaminants show a sharp rise which can hardly be explained by the modest decrease of the rainfall rate. It is as if air of different origin became involved in the precipitation process, though this is not reflected by the δ-measurements.

During the coldfront passage (19.00–22.00 G.M.T.) a hitherto unexplained anomaly occurs in the calcium data at 20.15 G.M.T. The δ-values show perhaps a slight response to the

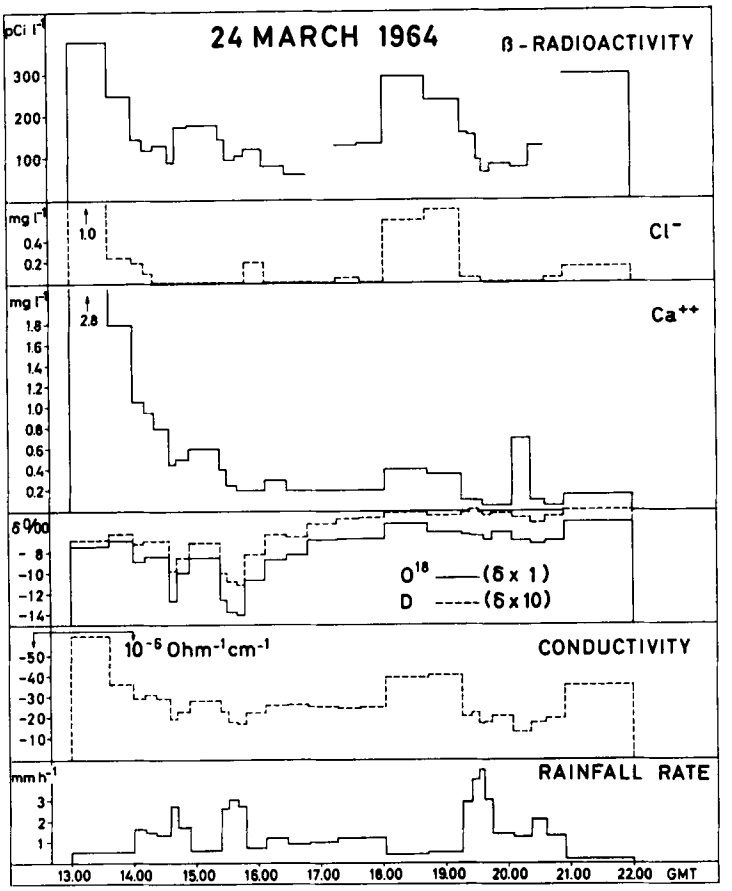

FIG. 7. Passage of a frontal system on 24 March 1964. Rainfall rate, conductivity, deuterium, oxygen-18, Cl^-, Ca^{++}, and specific radioactivity versus time.

coldfront pattern but less than in the other coldfront cases.

In Fig. 9 the δ_D for this rain has been plotted against the δ_{18}. The first nine samples represent the precipitation from the highest part of the warm front, i.e. the rain which was exposed to considerable evaporation during its fall through the relatively dry continental air below the frontal surface. The slope of the enveloping figure is less than 8, showing the evaporation to be fast and to take place under non-equilibrium conditions (cf. sub-paragraph 3.1).

Similar indications of non-equilibrium processes were not found in the other three cases where both heavy isotopes were measured (sub-paragrahs 4.1, 4.2 and 4.3). This could be due to these three cases occurring in late summer with higher humidity and, consequently, slower evaporation from falling drops.

5. Remarks

From the above it follows that with the aid of a very simple sampling method of precipitation some interesting facts can be established on the variability of the composition of the precipitation. It can however not be denied that the simplicity of the design of the experiment introduces a certain degree of speculation in the interpretation. This is why the authors consider the present research as a reconnaissance on the subject that might be of value for the design of a more sophisticated future research program in this field, that should include radar scanning of the cloud system, sampling of cloud droplets by aircraft, differentiating according to cloud droplet size, measurement of humidity and isotopic composition of the water vapour under the cloud system etc.

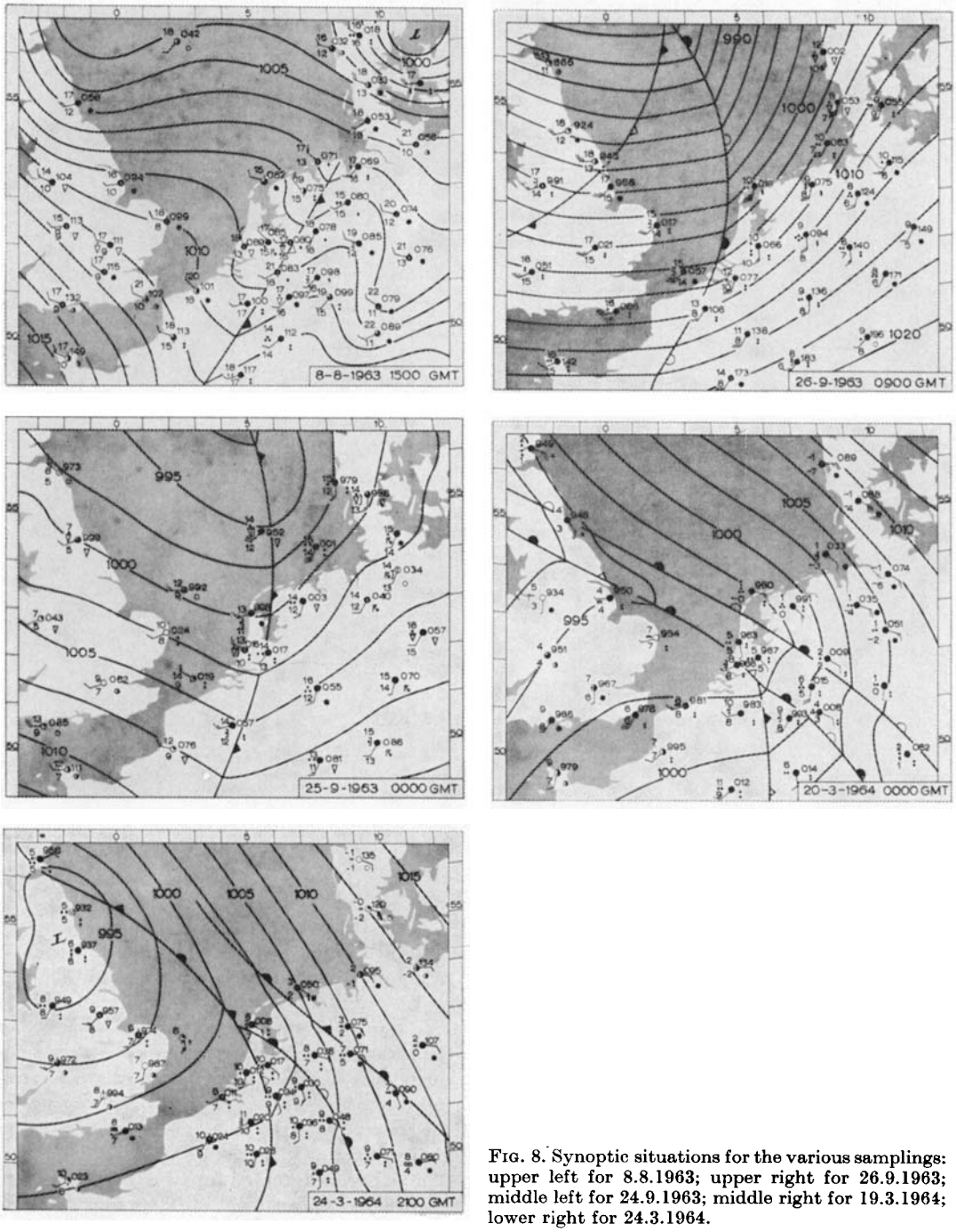

FIG. 8. Synoptic situations for the various samplings: upper left for 8.8.1963; upper right for 26.9.1963; middle left for 24.9.1963; middle right for 19.3.1964; lower right for 24.3.1964.

6. Acknowledgements

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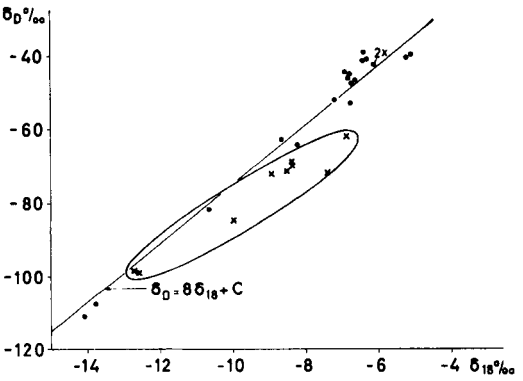

Fig. 9. δ_D versus δ_{18} for the rain of 24 March 1964. The first nine samples are indicated as ×.

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НЕКОТОРЫЕ ЗАМЕЧАНИЯ ОБ ОДНОВРЕМЕННЫХ ИЗМЕРЕНИЯХ ЗАГРЯЗНЕНИЙ В ВИДЕ ЧАСТИЦ И РАДИОАКТИВНОСТИ И ИЗОТОПНОГО СОСТАВА ОСАДКОВ

Обсуждаются результаты измерений радиоактивной пыли в дожде, собранной во время прохождения системы осадков. Образцы концентраций для загрязнений в виде частиц сопоставляются с пробами влаги по содержанию O_{18} и дейтерия. Изменения концентрации пыли исследованы для различных синоптических ситуаций.